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Decay fungi and certain other microorganisms use structural components of wood in their metabolism. Brown-rot fungi remove cellulose and hemicellulose from wood and white-rot fungi additionally use the lignin components of wood. Brown rot is characterized by rapid deterioration of the strength properties of wood already at an early stage of decay, before changes observable by the bare eye. For this reason, among others, brown-rot fungi are in the boreal climate zones the worst destroyers of wood and wood structures, causing annually losses of thousands of millions of Finnish marks.

Considerable drawbacks are associated with present-day wood preservatives. They contain wide-spectrum active agents, for which reason their adoption into use presupposes acceptance by public authorities. The action of the impregnants is based on so-called overall toxicity targeted at vital metabolic functions, such as cell breathing and the production of a high-energy compound, ATP, common to all living organisms. Since what is concerned is compounds with a wide-spectrum biologic effect, the use of present-day wood preservatives is associated with considerable health risks (e.g. carcinogens) and

environmental risks (pollution of soil and watercourses). Furthermore, the EU biocide directive now in force limits the use and development of biocidal compounds.

From FI patent publication 90951 there is known an alternative wood preservation method solving the problems associated with conventional preservation of sawn timber. In the said method, complexing agents are used for binding out of wood at least some of the metals essential for the metabolism of microorganisms. Wood preservation is carried out by impregnating the wood to be treated with a solution of a complexing agent, in a manner known *per se*.

10 It has been observed that the method effectively inhibits decay caused by decay fungi. Furthermore, a treatment with, for example, EDTA prevents the formation of damage caused by certain wood-destroying insects. However, many complexing agents, such as precisely EDTA, are associated with the disadvantage that the compound washes out.  
15 Efforts have been made to prevent this by various means, but usually with poor results.

The object of the present invention is to eliminate the disadvantages of prior art and to provide a novel method for protecting timber, such as sawn timber, wood composites such as plywood, chipboards and fiberboards, and similar lignocellulose-based products against  
20 rotting, molding and similar decay and damage reactions caused by microorganisms.

By the method according to the invention, the penetration of water into the wood structure can be limited by treating the wood with water-repellent compounds. Thus, the invention uses compounds such as siloxane derivatives, acid anhydride derivatives containing a  
25 hydrophobic carbon chain, or fluoroalkyl polymers, which are capable at least in part of penetrating also into the cell wall structure of wood and of preventing interaction between hydroxyl groups and water. It would appear that the hydrophobification compound binds covalently or polymerizes with the reactive compounds of the cell wall of a lignocellulose-based material, whereupon there forms in the surface structures of the material a water-  
30 repellent film which prevents the penetration of water molecules into the macrostructure of the lignocellulose-based material.

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It has been observed that the dimensional stability of wood is thereby improved. As the moisture content of wood decreases, the conditions for action and the effects of external factors, such as fungi, are weakened. The present invention aims at controlling the moisture content of lignocellulose-based materials in such a manner that the transfer of ambient moisture into the wood cell wall is prevented. It has been observed, unexpectedly, that a very good wood preservation effect is achieved by combining such a hydrophobification treatment with the above-mentioned wood preservation using a complexing agent as the active component (as a fungicide-like agent).

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The moisture content of the wood material is crucially important for the growth and propagation of microorganisms. Decay fungi require relatively high moisture contents in the substrate, water at minimum 30 % of the dry weight of the wood. This moisture content causes saturation of wood cell walls with water and the formation of a free water film in the cell cavity. The prevention of the transfer of water into the cell wall leads to a situation in which the moisture requirements of decay fungi are not fulfilled, and the growth and propagation of fungi are not possible. At the same time, hydrophobification agents are surprisingly effective in binding complexing agents to wood.

20 More specifically, the method according to the invention is mainly characterized in what is stated in the characterizing part of Claim 1.

The wood preservative composition according to the invention for its part is characterized in what is stated in the characterizing part of Claim 9, and the method according to the invention for controlling the moisture content of a lignocellulose-based material is characterized in what is stated in the characterizing part of Claim 11.

Considerable advantages are gained by means of the invention. Thus, by means of the invention the resistance of wood material to damage caused by microorganisms can be improved without the use of conventional fungicides, or the amount of fungicide required for inhibiting rot in wood can be lowered significantly.

The method according to the invention is suited for all lignocellulose-based products (e.g. sawn timber, plywood, chipboards, and various wood composites). The method according to the invention is used in a treatment combination aimed at improving the biologic efficacy of the treatment (e.g. EDTA + hydrophobification treatment). In principle, the hydrophobification treatment is also suited for decreasing the amount of fungicides (e.g. conventional impregnant + hydrophobification treatment). A lignocellulose-based material can be treated by various pressure impregnation methods, application and spraying methods, and immersion methods.

Water immersion/drying tests have shown that the hydrophilicity and dimensional instability of wood treated with combination solutions is clearly less than that of untreated wood. An EDTA addition would also seem to enhance the effect of siloxane, since the ASE values of specimens treated with combination solutions are many times the values obtained using a 5 % siloxane solution. When the cycling is repeated, the ASE values usually increase by degrees, which is often due to the fact that the dimensional instability of the control specimens increases strongly as cycling progresses.

When meeting the property requirements according to the invention, the compound improving water repellence penetrates into the cell wall and becomes there unwashable or difficult to wash out (a compound which either forms covalent bonds or is polymerizable). Furthermore, the compound is preferably emulsifiable in water.

The results obtained in connection with the invention give reason to assume that by the combining of a complexing agent and liquid siloxane (e.g. EDTA and SISW) it is possible to obtain a product competitive with the currently used wood preservative products, particularly those aimed at class AB.

The invention will be described below in greater detail with the help of a detailed description, with reference to the accompanying drawings, wherein

Figure 1 depicts the contact angles of water as a function of time on wood surfaces (surface parallel to the radius) treated with chelator-siloxane combination solutions;

Figure 2 depicts, with the help of a bar diagram, the differences in the dry volumes of the wood specimens before and after impregnation;

- 5 Figure 3 depicts, in the form of a bar diagram, the effect of compounds improving water repellence on the dimensional stability of wood;

Figure 4 depicts the decay preventing efficacy of a combination treatment against rot caused by *Poria placenta*;

- 10 Figure 5 depicts, with the help of a graph, the (washed) anti-rot efficacy of combination treatments;

Figure 6 depicts, with the help of a graph, the anti-fungus efficacy of chelator-siloxane combinations, and

Figure 7 depicts, with the help of a bar diagram, the anti-rot efficacy of the combination treatments in an earth pot test simulating ground contact.

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By “non-desirable reactions” of microorganisms is meant in the present applications primarily the damage and decay of wood caused by fungi and molds. The decay of wood, i.e. a substantial weakening of its strength properties, is mainly attributable to decay fungi, which include, for example, the above-mentioned brown-rot fungi and white-rot fungi.

- 20 Damage (i.e. discoloration) to wood is caused, among others, by the above-mentioned blue-stain fungi and mold fungi. However, the weight loss caused by these is insignificant.

The invention can be used for protecting timber from the non-desirable reactions of all of the above-mentioned microorganisms

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Within the framework of the present invention, the term “complexing agent” (i.e. “chelator”) denotes a substance capable of binding bi- or trivalent cations into insoluble or soluble complexes.

- 30 Complexing agents can be divided into inorganic and organic compounds. Inorganic complexing agents are various cyclic and linear phosphate compounds, for example,

- polyphosphates such as sodium polyphosphate ( $\text{Na}_5\text{P}_3\text{O}_{10}$ , STPP). The most important organic complexing agents are aminocarboxylic acids and their salts having acetic acid as the acid part (some examples to be mentioned are ethylenediaminetetra-acetic acid (EDTA), n-hydroxyethylethylenediaminetriacetic acid (HEDTA), diethylenetriaminepenta-
- 5 acetic acid (DTPA), nitroloacetic acid (NTA), ethylenediamine-di-(o-hydroxyphenylacetic acid) (EDDHA), diethanolglycine (DEG) and ethanoldiglycine (EDG), and salts thereof, in particular alkali metal salts, hydroxy acids (gluconic acid, glucoheptonic acid and other saccharic acids such as  $\beta$ -glucoisosaccharic acid,  $\alpha$ -isosaccharic acid, tartaric acid, maleic acid, and citric acid) and salts thereof, and organophosphates having phosphoric acid as the
- 10 acid part (examples to be mentioned include aminotrimethylenephosphonic acid (ATMP), 1-hydroxyethylidene-1,1-diphosphonic acid (HEDP), ethylenediaminetetramethylenephosphonic acid (EDTMP), diethylenetriaminepentamethylenephosphonic acid (DTPMP), and salts thereof.
- 15 In the invention it is also possible to use metal-binding phenolates or catecholates, such as biologic chelators, siderophores, produced by microorganisms. Siderophores are microorganism-produced complexing agents which bind metal ions of the substrate, in particular iron, for the use of the organism. It has been observed that certain siderophores produced by fungi (*Trichoderma sp.*) or bacteria (*Pseudomonas sp.*) have an effect
- 20 inhibiting the growth of other microorganisms, which is based on a strong affinity of siderophores to the iron in the substrate.

According to a preferred embodiment of the invention, the complexing agent used is aminocarboxylate or aminocarboxylic acid, most preferably aminotetracarboxylates or

25 aminotetracarboxylic acids. In the examples presented below there are used ethylenediaminetetra-acetic acid (EDTA) and salts thereof (ethylenediaminetetra-acetates), which have proven to be especially effective in the method according to the invention. The suitable salts of EDTA include alkali metal salts, in particular sodium salts:  $\text{Na}_2\text{-EDTAc}$  and  $\text{Na}_4\text{-EDTAc}$ .

In the following description, reference is made in particular to EDTA. It should be noted, however, that other complexing agents behave in a corresponding manner, and the invention is not limited specifically to EDTA.

- 5 In connection with the invention, wood material (includes wood, various wood products and wood composite products) is treated with hydrophobification compounds. The action of hydrophobification compounds is based on their molecular structure, wherein there is a hydrophilic end at one end of the molecule and a hydrophobic one at the other. The hydrophilic end of the compound favors an aqueous phase (e.g. the hydrophilic OH groups of the wood cell wall) and the hydrophobic end favors an oil phase, i.e. it repels water. Typical compounds described above include various siloxanes and their derivatives, acid anhydride derivatives containing a hydrophobic carbon chain, and various fluoroalkyl polymer derivatives (e.g. polytetrafluoroethylene). Examples to be mentioned of the usable silicon compounds (e.g. siloxane compounds) include those used in, for example, the textile industry, the concrete and masonry industry, and the paper industry (deinking and coating of paper) for hydrophobification purposes.
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- Suitable silicon polymers, fluorinated polymers, alkylketenedimers and acid anhydrides have been described, for example, in publications JP63176101, JP4070302, JP59033133, EP 0 747 183, WO80/02249, US 4.044.172, 4.404.306. The materials mentioned in these publications are incorporated into the present application by reference.
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- Particularly preferred compounds include siloxane or corresponding liquid silicon compounds such as polydimethylhydrogensiloxane, polymethylhydrogensiloxane, polydimethoxysiloxane, aminofunctional polydimethoxysiloxane, modified reactive polysiloxane, modified oligomeric siloxane, oligomeric siloxane, dimethylsiloxane, and phenylmethylpolysiloxane.
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- In this specification, reference is made in particular to the product SISW siloxane, which represents a preferred embodiment. The compound concerned illustrates generally the
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silicon compounds usable in the invention, and the intention is not to limit the invention to it.

In the present invention it has been shown that the decay preventing efficacy of EDTA and SISW can be improved with a mixture formulated from these active agents. The mixing of the compounds further results in that the required efficacy against decay is arrived at by using lower concentrations than if individual active agents were used.

Na<sub>2</sub>-EDTA and SiSW siloxane form a mixture with which sapwood of pine can easily be vacuum impregnated. In our tests we have observed that a mixture containing 1 % of Na<sub>2</sub>-EDTA and 3 % of SiSW siloxane is very effective against decay caused by *P. placenta*. Even after a leaching, the active agent mixture remains in the cellular system in an amount sufficient to prevent a weight loss (<3 % weight loss). A comparison of the efficacy against decay of a 1 % Na<sub>2</sub>-EDTA-3 % SISW mixture with the decay preventing efficacy of individual Na<sub>2</sub>-EDTA and SISW siloxane showed that the mixture enhances the efficacy of the individual active agents. At the concentration used in the test, neither active agent alone provides sufficient action against decay (weight loss less than 3 %) after leaching.

In general the amount of complexing agent is approx. 0.1 – 100 kg, preferably approx. 1 – 30 kg, especially preferably approx. 1.5 – 20 kg of complexing agent/m<sup>3</sup> of dry wood. The amount of a silicon/siloxane compound is within a corresponding range, i.e. approx. 0.1 – 100 kg/m<sup>3</sup> of dry wood. Wood or a corresponding lignocellulose-based material can be treated separately with a complexing agent and a hydrophobification agent, either first with a complexing agent and then with a hydrophobification agent, or *vice versa*, or the material can be treated with solutions of both agents simultaneously. The material may be dried between the treatments. Especially preferably a lignocellulose-based material is treated with a mixture of a complexing agent and a hydrophobification agent, the mixture containing 0.01 – 30, preferably approx. 0.1 – 20, especially preferably 0.5 – 10 % by weight of complexing agent and 0.01 – 40, preferably approx. 0.1 – 30, especially preferably 0.5 – 10 % by weight of hydrophobification agent.



Timber can be impregnated with an agent improving water-repellence by any method known *per se*, for example, by pressure, vacuum, vacuum+pressure impregnation, immersion treatment, application treatment or spray treatment. For example, according to one option, timber is impregnated with a hydrophobification compound in a vacuum of approx. 10 – 95 %, preferably approx. 70 – 90 % vacuum (duration of the treatment approx. 10 min – 5 h, preferably approx. 30 min – 2 h). Thereafter any excess treatment solution is removed, which can be carried out first at normal pressure and thereafter at lowered pressure, whereafter the pressure is raised to an overpressure of approx. 2 – 20 atm, preferably to a value of approx. 5 – 15 atm.

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Timber can be impregnated with a hydrophobification agent by an immersion treatment. The last-mentioned option can be carried out simply, for example, by immersing the timber to be treated (e.g. sawn timber) in a vat containing a hydrophobification agent. In the immersion treatment there is used a maximally saturated complexing agent solution, in which case the treatment time in the impregnation step is approx. 1 min – 5 h. The time required by the immersion treatment of fresh sawn timber is typically approx. 30 min – 2 h.

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The model compounds tested penetrate the cell wall and form therein, with the reactive compounds of the cell wall, stable combinations which do not washout or which wash out with difficulty. In the surface structures of wood material treated with hydrophobification compounds there is formed a water-repellent film, which slows down the penetration of water molecules into the macro-structure of the wood material. In other words, the lignocellulose-based material has a water-repellent film which entirely or in part prevents the passage of ambient moisture into the wood cell wall.

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When penetrating into the cell wall, the hydrophobification compounds cause swelling of the cell wall structures and combine with hydrophilic groups therein, whereupon ambient water molecules either will not have room in the cell wall structures or will not find free bonding surfaces. The moisture balance of the cell wall structures of a wood material treated with hydrophobification compounds drops to a level (water = 30 % of the dry weight

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of wood) which is insufficient for the growth and propagation of fungi. Thus the treatments also prevent the wood strength weakening caused by decay fungi.

The causes of the improved efficacy against decay of the EDTA-siloxane mixture are at present not known precisely. It is possible that the compounds react with each other, forming a complex which retains and also promotes those properties of the treatment required for decay preventing action.

The following non-limiting examples describe the invention in greater detail.

The following substances were used in the examples:

Siloxane: Perlit SISW (Bayer AG, polydimethyl hydrogen siloxane)

EDTA: Na<sub>2</sub>EDTA (Akzo, Basf)

#### EXAMPLE 1

*Efficacy of combination treatment in preventing the wetting of a wood surface*

The indicator used for water-repellence of a surface is the shape of a drop of water dropped on the surface concerned, i.e. the angle between the drop and the base (contact angle): the greater the angle, the more water-repellent the base. When wood is rendered water-repellent, the penetration of water into the cell structure of the wood is substantially slowed down.

Contact angle measurements were carried out on wood surfaces which had been treated with a siloxane derivative and a combination mixture of a chelator and siloxane. 5-percent (siloxane) and 3-percent (chelator) treatment solutions were prepared from the base solutions of the compounds. Specimens of pine sapwood (20 x 20 x 5 mm) were vacuum impregnated with the treatment solutions, whereafter the specimens were oven dried (103 °C, 24 h). The effect of the treatments on the water-repellence properties of the wood surface was investigated by determining the contact angles of distilled water on the radially

cut surfaces of the specimens. Before the measurements the specimens had been stabilized under a relative humidity of 65 %.

Figure 1 depicts the contact angles of water as a function of time on wood surfaces (radial surface) treated with chelator-siloxane combination solutions. The controls were wood treated with water and wood treated with a siloxane solution. The contact angle measurements showed that treatments with the chelator-siloxane combination improved the water-repellence of wood. The water-repellence values of the wood surface obtained with the combination treatment are better than the values obtained with a treatment with siloxane alone.

## Example 2

### *Penetration into wood cell wall of compounds in combination treatment*

The penetration of compounds into the cell wall of wood is indicated by expansion of the wood, i.e. a greater dry volume of the wood after impregnation than before impregnation. In order to determine the change in the dry volume of the wood, the precise dimensions of pine sapwood specimens (21 x 21 x 5 mm) were measured oven dry before the impregnations and, further, oven dry after the impregnations.

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Figure 2 shows the differences in the dry volumes of the wood specimens before and after impregnation. The dry volume of wood impregnated with water (control) is smaller after the impregnation than before the impregnation, i.e. substances are leached out from the cell wall.

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On the basis of the results, the siloxane-chelator mixture penetrates into the cell wall.

### Example 3

#### *Effect of combination treatment on dimensional stability of wood*

The interaction of the hydrophilic groups in the cell wall with molecules other than water molecules results in a decrease of bonding sites available to water molecules. The improvement of the dimensional stability of wood under the effect of impregnation treatments is an indication of the penetration, into the cell wall of the wood, of the compound used for the impregnation and, to a certain degree, also of its bonding thereto.

- 10 The effect of hydrophobification compounds on the hydrophilicity and dimensional stability of pine sapwood was investigated with tests known as water immersion/drying tests. Pine specimens (21 x 21 x 5 mm) were vacuum impregnated with siloxane and with a chelator-siloxane combination. After drying subsequent to the impregnation, the dimensions of the specimens were measured. Thereafter the wood specimens were vacuum
- 15 impregnated with water and the dimensions of the specimens were measured while they were wet and also after oven drying. Impregnation with water and drying, as well as the measurements, were repeated.

20 Dimensional stability is expressed using so-called ASE values, which are obtained from the following calculation formulae:

ASE (%) =  $100 \times (S_c - S_t) / S_c$ , where  $S_c$  is the volume swelling coefficient of the control sample and  $S_t$  is the volume swelling coefficient of the treated specimens.  $S(\%) = 100 \times (V_w - V_d) / V_d$ , where  $V_w$  is the volume of wood impregnated with water and  $V_d$  is the

25 volume of oven dry wood.

Positive ASE values are an indication that there is less hydrophilicity and dimensional instability in the treated specimens than in the control specimens.

In Figure 3, the effect of hydrophobification compounds on the dimensional stability of wood is examined. The dimensional stability of the specimens was calculated in 2-3 successive water immersion/drying cycles.

- 5 The water immersion/drying tests indicated that the hydrophilicity and dimensional instability of wood treated with the combination solutions is clearly less than that of untreated wood. The adding of a chelator would also appear to enhance the effect of siloxane, since the ASE values of specimens treated with the combination solutions were many times the values obtained with 5-percent siloxane solutions.

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#### Example 4

#### *Efficacy of combination treatment against decay caused by brown-rot fungus in accelerated test conditions according to standard EN 113*

- 15 Treatments with a combination made up of a metal chelator and a siloxane derivative were selected for treatments according to the invention.

Of the compounds being tested, 3- and 5-percent working solutions were prepared from siloxane, and in the combination the chelator content was 1-2 percent and the siloxane content ranged from 2.5 to 5 percent. Pine sapwood specimens (15 x 15 x 5 mm) were vacuum impregnated with impregnation solutions. After impregnation, some of the specimens were exposed to a washing test of 4 days. An accelerated rotting test, modified according to the EN 113 standard, was performed on the test specimens. The test fungus selected was brown-rot fungus *Poria placenta*. The rotting test lasted for 5 weeks.

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Figure 4 shows the anti-rot efficacy of the combination treatment against decay caused by *Poria placenta*. The concentration of chelator was 1 % in all of the mixtures used. The siloxane concentration ranged from 1 to 5 %. The controls in the test were untreated controls and specimens treated with 3-percent EDTA and with 5 % siloxane.

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Figure 5 shows with the help of a graph the anti-rot efficacy of combination treatments (washed). The graph indicates the active agent concentrations required for achieving the less than 3 % weight loss required by EN standards.

- 5 According to the decay test results, the weight loss limit (<3 %) according to the EN standard requirements against decay caused by *Poria placenta* was achieved with an active agent combination wherein the concentration of siloxane was 3 % and the concentration of chelator was 1 %. The amounts absorbed into the wood were in this case approx. 23 kg/m<sup>3</sup> for siloxane and approx. 7.5 kg/m<sup>3</sup> for chelator.

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#### Example 5

##### *Efficacy against mold of combination treatment*

- 15 The efficacy of the combination treatment against mold (antifungus efficacy) was tested in a laboratory by the suspension method. For the test, pine sapwood specimens were impregnated with chelator-siloxane combinations wherein the chelator concentration was 1.5 % or 3 %, and the siloxane concentration was 1 % or 2 %. In addition, test specimens were impregnated with chelator alone (1.5 % or 3 %) and with siloxane alone (1 % and 2 %).

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- The test and control specimens were suspended in a random order in incubation boxes. The relative humidity of the air in the boxes was adjusted by means of water to 95 – 100 %, the test temperature being 20 °C (+/- 2 °C). Before the beginning of the test, a mold fungus suspension was sprayed into the test boxes. The mold suspension contained three mold species thriving well in wood: *Aspergillus versicolor* (E1), *Gladosporium sphaerospermum* (R7) and *Penicillium* sp. (1017). The forming of growth on the surfaces of the test specimens was followed microscopically and visually at 2-week intervals for 10 weeks. The amount of the formed growth was assessed according to a scale of 0 – 5.

- 30 0 = no growth  
1 = indications of growth

- 2 = 1 - 10 % of the surface area covered by microbial growth  
 3 = 10 - 30 % of the surface area covered by microbial growth  
 4 = 30 - 70 % of the surface area covered by microbial growth  
 5 = >70 % of the surface area covered by microbial growth

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The efficacy of the chelator-siloxane combinations against mold is shown in accompanying Figure 6. The combinations used in the test contained 1.5 % or 3 % of EDTA and 1 % or 2 % of SISW siloxane. The controls in the test were untreated controls and specimens treated with 1.5 % or 3 % EDTA and with 1 % and 2 % SISW siloxane.

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The anti-fungus efficacy of the combination treatment is significantly better than the anti-fungus efficacy of either individual compound in the combination. Pine sapwood treated with the combination treatment did not mold at all during the 10-week exposure to mold.

#### 15 **Example 6**

##### *Efficacy of combination treatment in a soil block test simulating ground contact*

A compost soil mixture was prepared for a soil block test (accelerated and modified EN 807) simulating contact with ground. The water binding capacity and moisture content of the soil mixture was adjusted to comply with the EN 807 standard (60 % and 55 %).

Pine sapwood specimens (30 x 10 x 5 mm) were vacuum impregnated with a combination mixture wherein the chelator concentration was 3 % and 6 % and the siloxane concentration was 7.5 % and 15 %. The test specimens were rinsed in the normal manner (according to the standard EN 807) after the impregnation. The controls in the test were untreated pine sapwood specimens. The duration of the test was 20 weeks, whereafter the weight loss in the specimens was determined.

The decay preventing efficacy of the combination treatments in a soil block test simulating ground contact is depicted in Figure 7.

The results obtained from the soil block test simulating ground contact indicate that a combination treatment significantly improves the resistance of treated timber in ground contact conditions.

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